Results and perspectives for study of heavy and super-heavy nuclei and elements at IMP/CAS

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Abstract

In the past 30 years, synthesis of new isotopes and study of their decay properties have been remaining as high-priority research program at the Institute of Modern Physics, Chinese Academy of Sciences (IMP/CAS). Up to now, 34 new isotopes have been synthesized, 22 of which are heavy and super-heavy nuclei. In this paper, the techniques used and the results obtained at IMP/CAS for the study of heavy and super-heavy nuclei and elements are reviewed chronologically and the perspectives in the near future are introduced.

Keywords: α decay; Fusion and fusion-fission reactions; Super-heavy nuclei

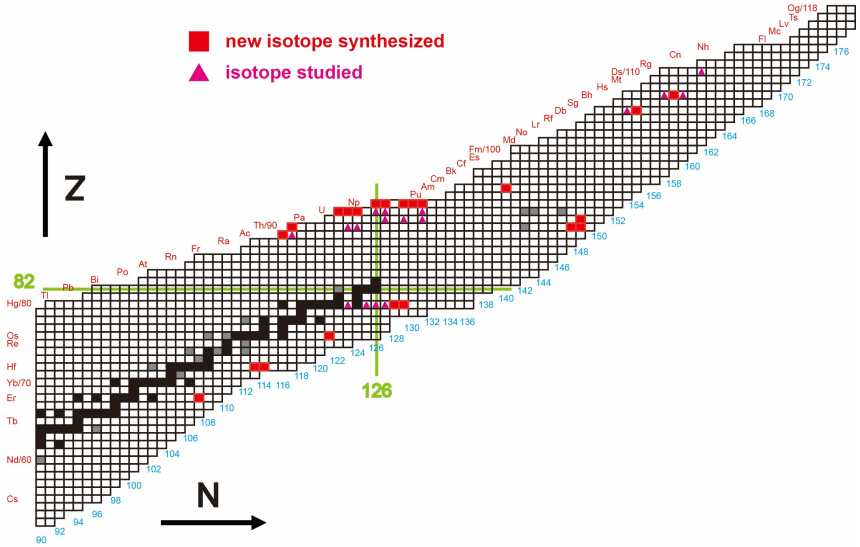

Fig. 1 Overview of heavy and super-heavy nuclei studied at IMP/CAS

1 Introduction

Due to the curiosity of the human beings on the unknown world, the synthesis and discovery of new elements to expand the Periodic Table of Elements becomes one of the frontier research fields of nuclear physics and chemistry. In the last 70 years, many laboratories around the world made a lot of efforts on this field to synthesize heavy and super-heavy nuclei/elements and study their physical and chemical properties. In China, such studies started in 1990s. Since then, a batch of new isotopes have been synthesized for the first time and their decay properties were studied experimentally. It becomes one of the high-priority research programs at the Institute of Modern Physics, Chinese Academy of Sciences (IMP/CAS). Up to now, 34 new isotopes have been synthesized, 22 of which are heavy and super-heavy nuclei. Figure 1 shows the location of these isotopes in the chart of nuclides.

Chronologically, these studies at IMP/CAS can be divided into three stages. During the early stage, the isotopes of interest were separated by using

radiochemical method and their decay properties were studied by using γ rays or β-γ/X-γ coincidences. Afterwards, helium-jet technique was developed and used to separate and transport the products away from the reaction sites, and their subsequent α, β and/or γ decays were detected. From 2010, a new gas-filled recoil separator was commissioned, and then the fusion-evaporation residues were transported and separated and the energy-position-time correlation method was used to assign the charge number Z and mass number A of the isotopes of interest.

In this paper, we will review the techniques used and the results obtained at IMP/CAS chronologically in Section 2 – 4 and give an introduction on the perspectives for the study of heavy and super-heavy nuclei and elements in the near future in Section 5.

2 The first stage

During the first stage, almost all the studies aimed at the neutron-rich heavy nuclei. The isotopes of interest were produced by multi-nucleon transfer reactions or neutron-induced reactions. The products were separated by using radiochemical method and their decay properties were studied by γ-rays or β-γ/X-γ coincidences.

2.1 ^{208,209}Hg

The neutron-rich $^{203,205-209}$Hg isotopes were synthesized and their decay properties studied by a ^{12}C or ^{18}O beam bombarding thick ^{nat}Pb targets at HIRFL (Heavy Ion Research Facility in Lanzhou) at IMP/CAS, and new isotopes ^{208}Hg and ^{209}Hg were synthesized for the first time. In the study of ^{208}Hg [1, 2], a high efficiency release, separation, and collection of Hg products was successful performed with a good selectivity by using a special offline gas phase

thermochromatographic process followed by a liquid-liquid procedure. The elemental assignment of mercury isotope was based on the identification of its β-decay daughter Tl observed in the periodically extracted Tl element sample growing in the separated Hg element product solution. The half-life of ^{208}Hg was measured to be 42^{+23}_{-12} min. Later, the device was upgraded and used online in the synthesis of the ^{209}Hg isotope [3]. Six neutron-rich mercury isotopes $^{203,205-209}$Hg were observed by a β-γ coincident measurement technique. The half-life of ^{209}Hg was determined to be 35^{+9}_{-6} s, and four γ rays following its β-decays were assigned. The half-life of ^{208}Hg was measured again and improved to be 41^{+5}_{-4} min.

2.2 ^{185,186}Hf, ^{237,238}Th and ^{239}Pa

The experiments searching for new isotopes ^{185}Hf [2, 4] and ^{237}Th [5] were carried out at the 300 kV Cockcroft-Walton accelerator at Lanzhou University and at the 600 kV machine at IMP/CAS using 14 MeV neutrons produced by reactions of deuterons with a rotating TiT (tritide titanium) target. Samples containing an average of 20 g of natural metallic tungsten powder and containing an average of 16 g of $(NH_4)_2U_2O_7$ powder were irradiated in the experiments searching for ^{185}Hf and ^{237}Th, respectively. The experiments for the other three new isotopes [6–8] were performed at HIRFL. The nuclei were produced by intermediate energy heavy ion (^{18}O) bombardments of thick natural uranium, or tungsten targets through multi-nucleon transfer reaction.

After the bombardments, the irradiated targets were rapidly transferred to a chemical laboratory by a pneumatic transport system. The irradiated targets were dissolved in hydrochloric acid solution or nitric acid and then the products of interest were separated radiochemically from the mixture of the

reaction products by different processes. Finally, the sources were prepared for γ counting by HPGe detectors.

The assignments were mainly based on the observation of the growth and decay of characteristic γ rays from the daughter nuclei. The half-lives of ^{185,186}Hf, ^{237,238}Th and ^{239}Pa were determined to be 3.5±0.6 min, 2.6±1.2 min, 5.0±0.9 min, 9.4±2.0 min, and 106±30 min, respectively.

2.3 ^{175}Er and ^{197}Os

The experiments searching for both new isotopes ^{175}Er [9] and ^{197}Os [10] were carried out at the 600 kV Cockcroft-Walton accelerator at IMP/CAS using 14 MeV neutrons, as mentioned in the previous section. Several natural ytterbium targets and platinum metal foils were used to produce ^{175}Er and ^{197}Os by (n,2p) reactions, respectively.

A rabbit system was used to quickly transport the irradiated samples into a well shielded lead chamber. A HPGe planar detector and two coaxial HPGe detectors or a clover detector consisting of four n-type coaxial germanium detectors were used for X-ray and γ-ray measurements, respectively. Energy and time spectra for the X and γ rays were recorded in both singles and coincidence modes.

The identification was made by the coincidence between the characteristics X and γ rays. The half-lives of ^{175}Er and ^{197}Os were determined to be 1.2±0.3 min and 2.8±0.6 min, respectively. Partial decay schemes were also proposed based on X-γ and γ-γ coincidence measurements.

3 The second stage and helium-jet period

During the second stage, the helium-jet technique was developed at IMP/CAS and extensively used for transporting the products away from the reaction

sites to reduce the background, essential for the measurements in the studies of neutron-deficient isotopes and heavy and super-heavy nuclei.

3.1 Experimental setups and detections

Beams of ^{22}Ne, ^{26}Mg, ^{32}S, ^{36}Ar, or ^{40}Ca with energies of 5–7 MeV/amu from HIRFL entered a target chamber filled with ∼1.1 atm helium gas, passing through a thick Havar window, and finally bombarded a self-supporting metal target, such as ^{58}Ni, ^{92}Mo, ^{96}Ru, ^{106}Cd, ^{112}Sn, or ^{241,243}Am. Reaction products recoiling out of the target foil were thermalized in the helium gas and attached themselves to the aerosol (NaCl, KCl or $PbCl_2$) clusters. Helium-jets then swept the clusters and reaction products through a Teflon capillary and implanted them into a rotating wheel, a movable tape, or a tantalum disk in a collection chamber.

Depending on the specific experiments, the collected samples were dealt with different procedures. In the studies of heavy and super-heavy isotopes, the samples were rotated away from the deposit place and stayed at front of a series of Si(Au) surface-barrier detectors, where the emitted α particles were detected, periodically. The precursors were identified by employing the correlation between the energy and time of the detected α decays. In the studies of light and medium-mass neutron-deficient isotopes, the tape station system was used. After a certain period of collection, the radioactivity on the tape was moved to a shielded counting chamber for the coincidence measurements between the energy and time of the detected protons, γ and/or X rays. While the first activity sample was being counted, the next one was being collected. Two fully depleted silicon surface barrier detectors with a thickness of 300 μm used for the proton measurements were located on two opposite sides of the movable tape. Behind each silicon detector, a coaxial HPGe detector was

used for measuring $\gamma(X)$ rays. The precursors were identified by employing the correlation between the β-delayed protons and the γ rays from the $2^+ \rightarrow 0^+$ states of the daughter nucleus. If the X ray was observed with good statistics, the assignment was made by the X-γ coincidences.

3.2 Super-heavy nuclei, ^{258,259}Db and ^{264,265,266}Bh

^{259}Db [11] and ^{258}Db were produced via the reaction ^{241}Am(^{22}Ne, 4-5n) at E_{lab}=118 MeV, while ^{266}Bh [12], ^{265}Bh [13] and ^{264}Bh via ^{243}Am(^{26}Mg, 3-5n) at E_{lab}=162 and 168 MeV. The reaction products were transported and collected using the helium-jet technique and the rotating wheel apparatus. The α-decays of the products and their daughter nuclides were detected by a set of Si(Au) detectors. The Z and A of the nuclide were unambiguously identified by the genetic relationship between the new activities and the known nuclides established by the recoil-α coincidence measurement.

The new nuclide ^{259}Db was determined to have a half-life of 0.51±0.16 s and decay by α-particle emission with E_α=9.47 MeV. The nuclide ^{258}Db has a half-life of 4.3±1.1 s and decays by α-particle emission with E_α = 9.08, 9.17 and 9.30 MeV, which are in agreement with previous known data [14], thus also proving the reliability of our assignment of ^{259}Db.

A total of 8 correlated decay events of the new isotope ^{265}Bh and 4 decay events of the known isotope ^{264}Bh were observed. ^{265}Bh decays with a half-life of $0.94^{+0.70}_{-0.31}$ s by emission of α particles with an average energy of 9.24±0.05 MeV. For ^{266}Bh, a total of 4 correlated decay events was observed. The measured α-particle energy and half-life for ^{266}Bh are 9.03±0.08 MeV and $0.66^{+0.59}_{-0.26}$ s, respectively. The E_α value is close to the 9.07 MeV for ^{266}Bh observed in the first chain of element 113 at RIKEN, providing supports the elemental assignment of the element 113 [15].

The isothermal gas chromatographic behaviour of the group 5 elements Nb, Ta and Db was also investigated [16] in a brominating atmosphere using the OLGA (On-Line Gas chromatography Apparatus) technique. It was found that Db forms a very volatile compound, most likely the pentabromide, being more volatile than similar compounds formed under identical conditions with Nb and Ta, respectively.

3.3 Light and medium-mass neutron-deficient nuclei

By utilizing the p-γ coincidence technique in combination with the helium-jet tape transport system, the β-delayed proton decays of nine new nuclides in the rare-earth region near the proton drip line (^{121}Ce [17], ^{125}Nd [18], ^{128}Pm [18], ^{129}Sm [18], ^{135}Gd [19], ^{137}Gd [18], ^{139}Dy [18], ^{142}Ho [20], and ^{149}Yb [21]) and five nuclides in the A=90 region with N$\sim$Z (^{81}Zr [22–24], ^{85}Mo [22–24], ^{89}Ru [25], ^{92}Rh [26], and ^{93}Pd [26]) were observed. The new nuclides ^{129}Pm [27] and ^{139}Tb [28] were studied by utilizing the X-γ coincidence technique with the helium-jet tape transport system. Ref. [29] reviewed these studies. In addition, ^{69}Kr [30, 31] was studied by using pulsed-beam technique in combination with a ΔE-E-E_{reject} particle telescope.

3.4 ^{235}Am

The new isotope ^{235}Am [32] was produced by the reaction ^{238}Pu(p,4n)^{235}Am using 35 MeV protons delivered by the proton linear accelerator at the Institute of High Energy Physics, CAS. A helium-jet multiple-target chamber system was used to transport the reaction products to a low-background area where the products were collected on a tantalum disc. The collected products on the tantalum disc were dissolved in nitric acid, and then the americium isotopes were chemically separated from fission products and other actinides by a rapid

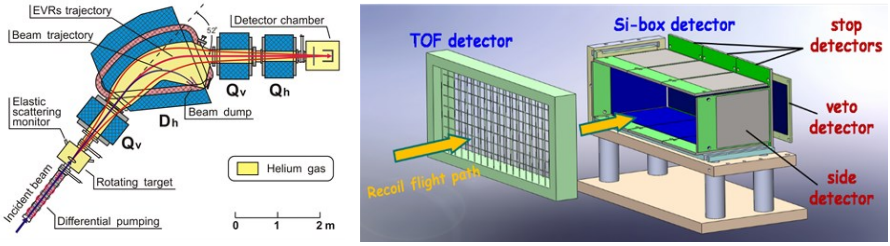

Fig. 2 Left: A schematic view of the gas-filled recoil separator SHANS. Right: The detection system placed at the focal plane.

procedure. From the observed KX rays of Np, γ rays of ^{235}Pu and X-γ coincidence measurements, the new isotope ^{235}Am with a half-life of 15±5 min was identified.

4 The third stage and gas-filled recoil separator SHANS period

Starting from 2010, all the studies about the heavy and super-heavy nuclei were performed at the new gas-filled recoil separator, SHANS (Spectrometer for Heavy Atoms and Nuclear Structure).

4.1 SHANS and detection

SHANS [33] is a gas-filled recoil separator with the type of $Q_vD_hQ_vQ_h$ configuration, where Q and D denote the quadrupole and dipole magnets, and the subscripts v and h refer to vertical and horizontal focusing direction, respectively. Its total length is 6.5 m and the angular acceptance is 25 msr. The maximum magnetic rigidity of the dipole magnet is 2.88 Tm and its bending angle is 52°. A silicon semiconductor detector box (Si-box) is installed at the focal plane of the separator. Figure 2 shows the layout of the SHANS and the detection system placed at the focal plane.

Three 300-μm thick position sensitive silicon detectors (PSSD) are mounted at the back of the Si-box as stop detectors. They are used for measuring implantation, fission fragments and α particles. Eight non-position-sensitive silicon detectors are mounted in an open box arrangement around the strip detectors. They are used to detect the escaping radioactive decay events. Three punch-through detectors are mounted behind the PSSD to provide veto signals for light particles passing through the strip detectors. A multi-wire proportional counter (MWPC) is mounted upstream of the Si-box as a timing detector. The time-of-flight (TOF) between the MWPC and PSSD allows us to distinguish the radioactive decay events in the PSSD from the implantation ones. The event chains consisting of implanted evaporation residues (EVR) and their subsequent α decay and/or spontaneous fission are identified by the position-time correlation method. The digital signal processing technique consisting of 16 14-bit flash Analog to Digital Converters (ADCs) from CAEN Costruzioni Apparecchiature Elettroniche Nucleari S.p.A. [34] has been implemented in the data acquisition system. The shapes of the preamplifier signals from all the detectors were independently recorded in traces with tens of microsecond long and 100-MHz sampling rate. The rise time of the preamplifiers was about 40 ns, which enabled a resolving fast decay signals following the implantation of EVRs.

4.2 ^{271}Ds

The super-heavy nuclide ^{271}Ds [35] was synthesized via ^{208}Pb(^{64}Ni,n) reaction at a beam energy of 313.3 MeV and separated and identified by SHANS just after its commissioning in 2011. Based on the separator coupled with a position sensitive silicon strip detector, the energy-position-time correlation measurements for the implanted nucleus and its subsequent decay α particles

were carried out. One α-decay chain for ^{271}Ds was established. The α-particle energy and decay time of the ^{271}Ds nucleus were measured to be 10.644 MeV and 96.8 ms, respectively, which are consistent with the values reported in the literature [36, 37].

The nucleus ^{271}Ds is the heaviest isotope synthesized and studied at IMP/CAS and also in China up to now.

4.3 Neutron-deficient actinides

With the continuous development and improvement of SHANS and related devices, 10 new isotopes in the most neutron-deficient actinide region from Ac to Np were synthesized for the first time and their decay properties were studied.

A lot of mass models (e.g. the macroscopic–microscopic models FRDM [38], Duflo-Zuker [39], WS4 [40], the Hartree–Fock–Bogoliubov models HFB-24 [41, 42], UNEDF0 [43], and the spherical relativistic continuum Hartree–Bogoliubov (RCHB) theory [44]) predict the existence of multiple neutron-deficient actinide isotopes which have not been found experimentally. Searching for new actinides, especially U and Np isotopes, around the proton drip line for the heaviest elements and then studying their decay properties are our motivations for these studies. In addition, these isotopes lie in the region with the magic number of N=126 and thus investigating shell structure evolution of the N=126 shell closure is highly intriguing. These studies are quite challenging due to the low production cross sections of these nuclei and their short half-lives.

Table 1 lists experimental details of these studies, in which the reaction systems and the corresponding production cross sections are included. By using fusion-evaporation reactions between beams of ^{36}Ar, ^{40}Ar and ^{40}Ca and

Table 1 Experimental details of neutron-deficient actinides studied at SHANS. E_b is the beam energy and σ the production cross section. TBP means "To be published".

Nuclide	Beam	Target	Channel	E_b (MeV)	σ	Ref.
^{204}Ac	^{40}Ca	^{169}Tm	5n			TBP
^{205}Ac	^{40}Ca	^{169}Tm	4n	196	∼70 pb	[45]
^{206}Ac	^{40}Ca	^{169}Tm	3n	183	∼0.3 nb	[45]
^{207}Th	^{36}Ar	^{176}Hf	5n	197–199	4^{+9}_{-3} pb	[46]
^{213m}Th	^{40}Ar	^{176}Hf	3n	183, 190		[47]
^{214}Th	^{40}Ar	^{176}Hf	2n	183, 190		[47]
^{218m}Pa	^{40}Ar	^{182}W	1p3n	190		[48]
^{220}Pa	^{40}Ar	^{187}Re	α3n	188, 198.7	$6.5^{+0.9}_{-0.7}$ nb at 188	[49, 50]
^{222}Pa	^{40}Ar	^{186}W	1p3n	198.7		[51]
^{214}U	^{36}Ar	^{182}W	4n	184	10^{+14}_{-7} pb	[52]
^{215}U	^{40}Ar	^{180}W	5n	204.5, 207.6	50^{+70}_{-30} pb at 207.6	[53]
^{216}U	^{40}Ar	^{180}W	4n	189.5, 191	300^{+170}_{-120} pb at 189.5	[52, 54]
^{218}U	^{40}Ar	^{182}W	4n	190	768^{+91}_{-82} pb	[52]
	^{40}Ca	^{184}W	α2n	206	325^{+47}_{-42} pb	[52]
^{219}U	^{40}Ar	^{183}W	4n	190	2.5±0.4 nb	[55]
^{223}U	^{40}Ar	^{187}Re	p3n	188		[56]
^{219}Np	^{36}Ar	^{187}Re	4n	191.5	19^{+44}_{-16} pb	[57]
^{220}Np	^{40}Ar	^{185}Re	5n	200	42^{+20}_{-15} pb	[58]
^{222}Np	^{40}Ar	^{187}Re	5n	198.7		[59]
^{223}Np	^{40}Ar	^{187}Re	4n	188	$0.9^{+0.3}_{-0.2}$ nb	[60]
^{224}Np	^{40}Ar	^{187}Re	3n	188	380^{+260}_{-110} pb	[61]

enriched isotopic targets of ^{169}Tm, ^{176}Hf, ^{180,182,183,184,186}W and ^{185,187}Re at different beam energies, nuclides from Ac to Np have been synthesized and the decay properties studied experimentally.

For the Np isotopes, the new nuclei $^{219,220,222-224}$Np have been synthesized and the following prominent results have been obtained. Firstly, according to the measured α-decay energy of ^{219}Np [57] and the masses of ^{215}Pa and ^{218}U [62] a proton separation energy S_p=−301±83 keV is determined and the proton drip-line for Np has been reached experimentally. Thus, in the region of nuclei with Z≥83, the proton drip line has been reached for all odd-Z isotopes up to

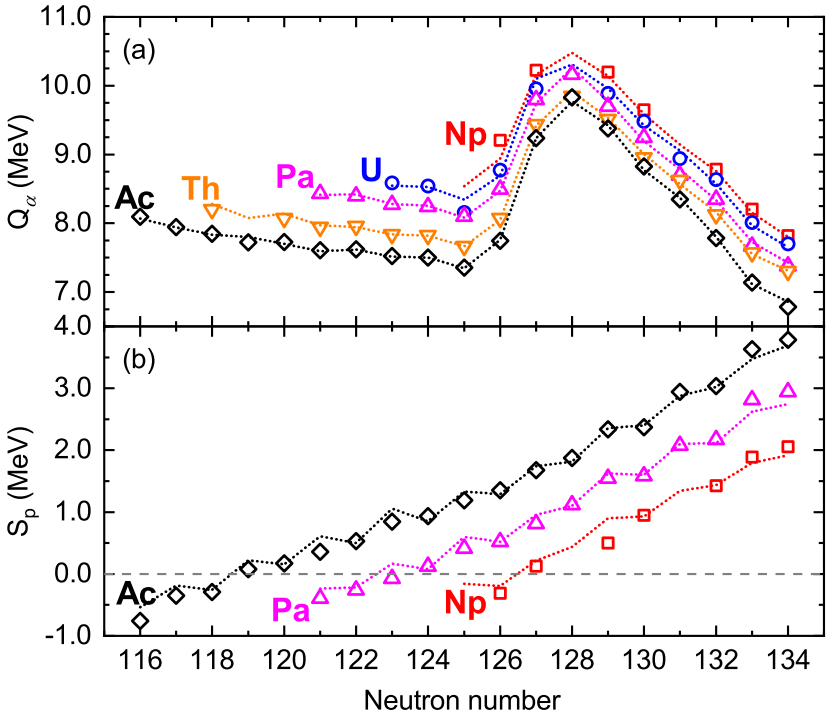

Fig. 3 Systematics of (a) α-decay Q_α values of ground-state to ground-state transitions for neutron-deficient $89 \leq Z \leq 93$ isotopes, and (b) proton separation energies S_p for odd-Z Ac, Pa, and Np isotopes as a function of neutron number.

Np. Secondly, by applying the digital signal processing technique, the half-life of ^{222}Np is determined to be only 380^{+260}_{-110} ns [59]. As the time of flight of the ^{222}Np EVRs through SHANS was estimated to be ~1.2 μs, this sub-μs value may be the shortest half-life which can be measured by SHANS. Thirdly, from the new experimental results the α-decay systematics for Np isotopes around N=126 is established, which allows us for the first time to test the robustness of the N=126 shell closure in Z=93 Np isotopes. By inspecting the systematics of the α-decay Q_α values, half-lives and reduced widths, the persistent rigidity of the N=126 magic number in Np isotopes is clearly recognized in our study [58]. Figure 3 shows the systematics of Q_α values of ground-state to ground-state transitions for neutron-deficient $89 \leq Z \leq 93$ isotopes, and proton separation energies S_p for odd-Z Ac, Pa, and Np isotopes as a function of neutron number.

The new isotopes of uranium ^{214,215,216}U [52–54] have been synthesized. By combining the experimental data, improved α-decay reduced widths δ^2 for the even-even Po–Pu nuclei in the vicinity of the magic neutron number N=126 are deduced. In order to study the influence of proton-neutron interaction on α decay in this region of nuclei, their systematic trends are discussed in terms of the N_pN_n scheme. It is strikingly found that the reduced widths of ^{214,216}U are significantly enhanced by a factor of two as compared with the N_pN_n systematics for the $84 \leq Z \leq 90$ and $N < 126$ even-even nuclei [52]. Figure 4 shows the systematics of reduced widths for ground-state to ground-state α decays of even-even $84 \leq Z \leq 94$ isotopes as a function of neutron number and against N_pN_n for even-even Po to U isotopes. The abnormal enhancement is interpreted by the strong monopole interaction between the valence protons and neutrons occupying the $\pi 1f_{7/2}$ and $\nu 1f_{5/2}$ spin-orbit partner orbits, which is supported by the large-scale shell model calculation.

5 Perspectives

In the near future, our studies on the heavy and super-heavy nuclei/elements can be performed on two facilities. Based on the existing gas-filled recoil separator SHANS, we will continue the studies on the neutron-deficient isotopes in the region of actinides and/or super-heavy nuclei, such as to synthesize new isotopes of Pu and Am and study their decay properties. At present, a new facility named CAFE2 including SHANS2 are being constructed. Synthesis of super-heavy elements and super-heavy nuclei is our long pursuing goal.

CAFE2 (China Accelerator Facility for super-heavy Elements) is a new facility constructed at IMP/CAS. It is based on an upgraded superconducting linear accelerator. A new gas-filled recoil separator SHANS2 (Spectrometer for Heavy Atoms and Nuclear Structure-2) [63] with a total flight path of 5.85

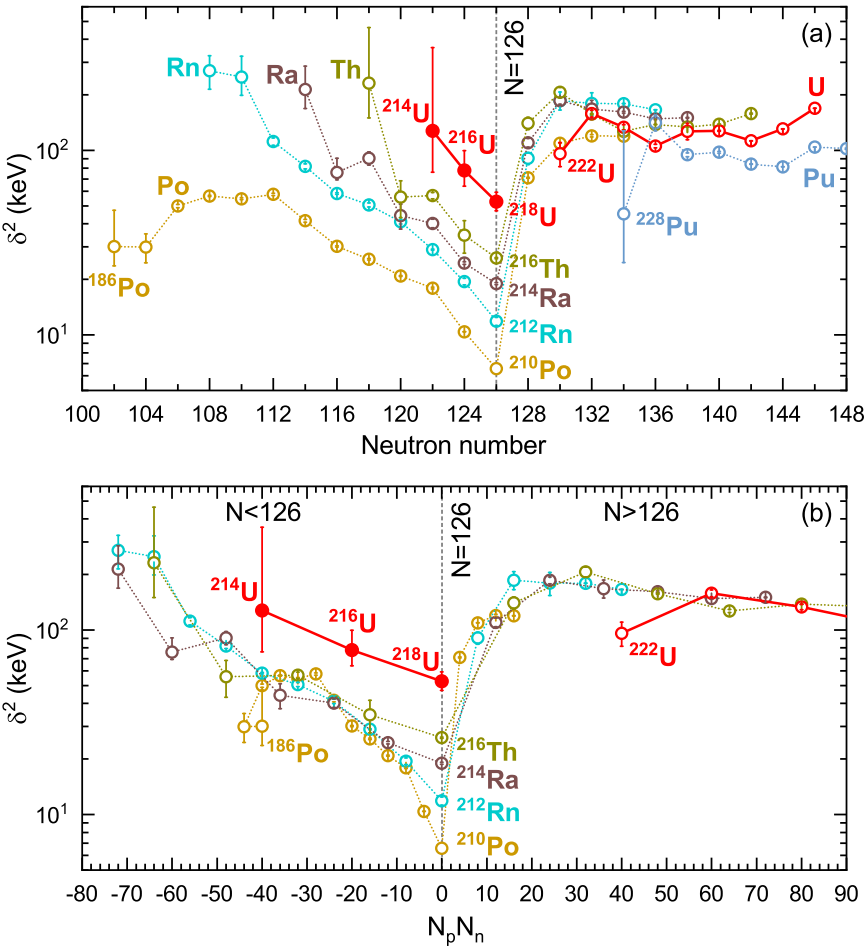

Fig. 4 (a) Systematics of reduced widths δ^2 for ground-state to ground-state α decays of even-even $84 \leq Z \leq 94$ isotopes as a function of neutron number. (b) Same as (a) but against N_pN_n for even-even Po to U isotopes. The N_p and N_n values are calculated relative to $Z = 82$ and $N = 126$ closed shells, respectively, with an exception of $^{186}_{84}Po_{102}$, for which $N_n = -20$, relative to the closest $N = 82$ neutron shell. From Ref. [52].

m at CAFE2 has an ion-optical configuration of $Q_vDQ_hQ_vD$. The maximum magnetic rigidity is 2.5 Tm and the maximum acceptable divergence angles of SHANS2 are ±70 mrad and ±113 mrad in horizontal and vertical directions, respectively. The techniques for the target, differential pumping, recoil detection and isotopic identification used successfully on SHANS will also be applied on SHANS2.

After the commissioning of CAFE2 and SHANS2, the efforts will be undertaken to produce and synthesize elements 119 and 120. Beams with medium mass, such as ^{50}Ti, ^{51}V, ^{54}Cr and ^{55}Mn, will be used to impinge on the radioactive actinide targets, such as ^{243}Am and ^{248}Cm. Because CAFE2 is a stand-alone facility devoted to such a study, a sufficient beam time would be expected, thus it gives us an opportunity to attempt to measure new elements.

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